

Morris-Shore transformation with unequal detunings

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The aim of this paper is to analyze the Morris-Shore (MS) transformation in the case of weak deviation of the condition for equal detunings. For this generalization of the MS transformation a perturbative solution is derived. Some elements concerning the general solution of the MS transformation are also discussed.

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I. INTRODUCTION

The dynamics of a two-state quantum system resulting from the action of a resonant external pulse of precise temporal area is a classical example in quantum mechanics. This simple two-state quantum system has an interesting theoretical extension, from a nondegenerate two-level quantum system (where only two quantum states /ground and excited/ are available), to one with a degenerate ground level and a degenerate excited level. These physical systems can be found at isolated atoms and molecules, where the states of well defined angular momentum J , due to the rotational symmetry produces a degeneracy of $2J+1$ magnetic sublevels. For correct description of laser-induced transitions between quantum states with defined angular momentum, one must also incorporate several magnetic sublevels (labeled by M), that may serve as a possible initial states. Hence each of these magnetic sublevels have a laser-driven excitation dynamic into to set of excited magnetic sublevels. Other source of multilevel systems can be the systems, whose mathematical description uses the usual rotating-wave approximation (RWA). Within the RWA the degeneracies can also be found for more general multistate quantum systems.

To handle such multilevel systems, in 1983 Morris and Shore have introduced a coordinate transformation with the general property that could factorize the dynamic of any two degenerate sets of quantum states, to an equivalent description involving only independent uncoupled pairs of equations. Generally, the Morris-Shore (MS) transformation is able to reduce the coherent quantum dynamics of a coupled degenerate two-level system to a set of independent nondegenerate two-state systems and a set of uncoupled states. The mathematical description of MS transformation only requires deriving of eigenvalues and the eigenstates of a hermitean matrix, which is a product of interaction matrices. The eigenstates of this hermitean matrix are the MS states, that represent the independent two-state systems and the dark states. The eigenvalues represent the MS interactions in the independent MS two-state systems. The MS transformation is quite general, but requires that all initial interactions are constant, or share the same time dependence, and also that all interactions are resonant, or equally detuned

from the upper states. The condition of equally detuned set of states implies that the lower set of states is degenerate in RWA sense, and so is the upper set of states.

Having explain the constraints of MS transformation, it is natural to ask whether they can be relaxed. It turns out that within the framework of weak deviation of equally detuned set of states, perturbative solution can be derived.

This paper is organized as follows. In Sec. II we review the two-level MS transformation and in details define the problem for its extension. In Sec. III we describe the perturbative solution to the problem of generalized MS transformation. Section IV presents an examples for the generalized MS transformation. Finally, Sec. V presents a summary of the results.

II. DEFINITION OF THE PROBLEM

We consider a two-level quantum system in the general case when both levels are degenerate. The ground **a** level consists of N_a degenerate states, the excited **b** level – of N_b degenerate states and we assume, without any loss of generality, that $N_a \geq N_b$. Every ground state $|\psi_i\rangle$ ($i = 1, 2, \dots, N_a$) is coupled via all excited states $|\psi_j\rangle$ ($j = N_a + 1, N_a + 2, \dots, N_a + N_b$) with pulsed interactions $\Omega_{ij}(t)$, each pair of which are on two-photon resonance, as depicted in ???. The excited level may be off single photon resonance by some detuning $\Delta(t)$ which should be the same for all couplings. We assume that the couplings share same time dependence $f(t)$

$$\Omega_{ij}(t) = \chi_{ij} e^{-i\xi t} f(t),$$

but we allow for different amplitudes χ_{ij} and thereby pulse areas.

The dynamics of the system is governed by the time-dependent Schrödinger equation which in the usual rotating-wave approximation (RWA) has the form

$$i\hbar \frac{d}{dt} \mathbf{C}(t) = \mathbf{H}(t) \mathbf{C}(t), \quad (1)$$

where the elements of the $(N_a + N_b)$ -dimensional state vector $\mathbf{C}(t)$ are the probability amplitudes of the states

and the Hamiltonian in a block-matrix form is given by

$$\mathbf{H}_0(t) = \begin{bmatrix} \mathbf{0}_{N_a \times N_a} & \mathbf{V}(t)_{N_a \times N_b} \\ \mathbf{V}^\dagger(t)_{N_b \times N_a} & \mathbf{\Delta}(t)_{N_b \times N_b} \end{bmatrix}. \quad (2)$$

Here $\mathbf{0}$ is the N_a -dimensional square zero matrix, in which the zero off-diagonal elements indicate the absence of couplings between the \mathbf{a} states, while the zero diagonal elements show that the \mathbf{a} states have the same energy, which is taken as the zero of the energy scale. The matrix $\mathbf{\Delta}(t)$ is an N_b -dimensional square diagonal matrix, with $\delta(t)$ on the diagonal, $\mathbf{\Delta}(t) = \delta(t)\mathbf{1}_{N_b}$. In Eq.(2) $\mathbf{V}(t)$ is an $(N_a \times N_b)$ -dimensional matrix, which elements are the couplings between the ground and the excited states, $\mathbf{V}^\dagger(t)$ is its hermitian conjugate

$$\mathbf{V}(t) = \begin{bmatrix} \Omega_{11} & \Omega_{12} & \cdots & \Omega_{1N_b} \\ \Omega_{21} & \Omega_{22} & \cdots & \Omega_{2N_b} \\ \vdots & \vdots & \ddots & \vdots \\ \Omega_{N_a 1} & \Omega_{N_a 2} & \cdots & \Omega_{N_a N_b} \end{bmatrix} \quad (3)$$

$$= [|\Omega_1\rangle, |\Omega_2\rangle, \dots, |\Omega_{N_b}\rangle], \quad (4)$$

where $|\Omega_n\rangle$ ($n = 1, 2, \dots, N_b$) are N_b -dimensional vectors, composed of the interaction energies of the n th state of the \mathbf{b} set with all states of the \mathbf{a} set,

$$|\Omega_n\rangle = \begin{bmatrix} \Omega_{1n} \\ \Omega_{2n} \\ \vdots \\ \Omega_{N_a n} \end{bmatrix}. \quad (5)$$

If the Hamiltonian describes the atom-laser interaction, in the presence of weak magnetic field the degeneracy of the states is lifted. Due to the external field we have an extra terms, which can be assumed as a perturbation to the exact MS solution. In this case we have $\mathbf{H}(t) = \mathbf{H}_0(t) + \mathbf{D}(t)$, where $\mathbf{D}(t)$ is diagonal matrix with dimension $N_a + N_b$ and generally $\mathbf{D}(t) \neq d(t)\mathbf{1}_{N_b}$. The aim of this paper is to analyze the MS transformation in the case of weak deviation of the $\mathbf{\Delta}(t) = \delta(t)\mathbf{1}_{N_b}$ condition, which is necessary for factorization towards set of two-state systems and set of decouple states. Some elements concerning the general solution of the MS transformation will be also discussed.

We will start our discussion by a brief description of the exact Morris-Shore (MS) transformation.

III. EXACT ANALYTICAL SOLUTION

A. Morris-Shore transformation

Morris and Shore [?] have shown that any degenerate two-level system, in which all couplings share the same time dependence and the same detuning, can be reduced

with a constant unitary transformation \mathbf{S} to an equivalent system comprising only independent two-state systems and uncoupled (dark) states, as shown in Fig. ???. This time-independent transformation is given by

$$|\psi_i\rangle = \sum_k S_{ik}^* |\tilde{\psi}_k\rangle \iff |\tilde{\psi}_k\rangle = \sum_i S_{ik} |\psi_i\rangle, \quad (6)$$

where the tildas denote the MS basis hereafter. The constant transformation matrix \mathbf{S} can be represented in the block-matrix form

$$\mathbf{S} = \begin{bmatrix} \mathbf{A} & \mathbf{O} \\ \mathbf{O} & \mathbf{B} \end{bmatrix}, \quad (7)$$

where \mathbf{A} is a unitary N_a -dimensional square matrix and \mathbf{B} is a unitary N_b -dimensional square matrix, $\mathbf{A}\mathbf{A}^\dagger = \mathbf{A}^\dagger\mathbf{A} = \mathbf{1}_{N_a}$ and $\mathbf{B}\mathbf{B}^\dagger = \mathbf{B}^\dagger\mathbf{B} = \mathbf{1}_{N_b}$. The constant matrices \mathbf{A} and \mathbf{B} mix only sublevels of a given level: \mathbf{A} mixes the a sublevels and \mathbf{B} mixes the b sublevels. The transformed MS Hamiltonian has the form

$$\tilde{\mathbf{H}}(t) = \mathbf{S}\mathbf{H}(t)\mathbf{S}^\dagger = \begin{bmatrix} \mathbf{0} & \tilde{\mathbf{V}} \\ \tilde{\mathbf{V}}^\dagger & \mathbf{\Delta}(t) \end{bmatrix}, \quad (8)$$

where

$$\tilde{\mathbf{V}} = \mathbf{A}\mathbf{V}\mathbf{B}^\dagger. \quad (9)$$

The $N_a \times N_b$ matrix $\tilde{\mathbf{V}}$ may have $N_d = N_a - N_b$ null rows (if $N_a > N_b$), which correspond to decoupled states. The decomposition of \mathbf{H} into a set of independent two-state systems requires that, after removing the null rows, $\tilde{\mathbf{V}}$ reduces to a N_b -dimensional diagonal matrix; let us denote its diagonal elements by λ_n ($n = 1, 2, \dots, N_b$).

It follows from Eq. (9) that

$$\tilde{\mathbf{V}}\tilde{\mathbf{V}}^\dagger = \mathbf{A}\mathbf{V}\mathbf{V}^\dagger\mathbf{A}^\dagger, \quad (10a)$$

$$\tilde{\mathbf{V}}^\dagger\tilde{\mathbf{V}} = \mathbf{B}\mathbf{V}^\dagger\mathbf{V}\mathbf{B}^\dagger. \quad (10b)$$

Hence \mathbf{A} and \mathbf{B} are defined by the condition that they diagonalize $\mathbf{V}\mathbf{V}^\dagger$ and $\mathbf{V}^\dagger\mathbf{V}$, respectively. It is important to note that the square matrices $\mathbf{V}\mathbf{V}^\dagger$ and $\mathbf{V}^\dagger\mathbf{V}$ have different dimensions, N_a and N_b , respectively. Because all elements of \mathbf{V} are constant, \mathbf{A} and \mathbf{B} are also constant; the elements λ_n are constant too. It is straightforward to show that the N_b eigenvalues of $\mathbf{V}^\dagger\mathbf{V}$ are all non-negative; according to Eqs. (9) and (10) they are λ_n^2 ($n = 1, 2, \dots, N_b$). The matrix $\mathbf{V}\mathbf{V}^\dagger$ has the same eigenvalues and additional $N_d = N_a - N_b$ zero eigenvalues.

The MS Hamiltonian (8) has the explicit form

$$\tilde{\mathbf{H}} = \begin{bmatrix} \mathbf{0}_{N_d} & \mathbf{0} & & & & & & & \\ & 0 & 0 & \cdots & 0 & \lambda_1 & 0 & \cdots & 0 \\ & 0 & 0 & \cdots & 0 & 0 & \lambda_2 & \cdots & 0 \\ & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ \mathbf{0} & 0 & 0 & \cdots & 0 & 0 & 0 & \cdots & \lambda_{N_b} \\ & \lambda_1 & 0 & \cdots & 0 & \Delta & 0 & \cdots & 0 \\ & 0 & \lambda_2 & \cdots & 0 & 0 & \Delta & \cdots & 0 \\ & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ & 0 & 0 & \cdots & \lambda_{N_b} & 0 & 0 & \cdots & \Delta \end{bmatrix}. \quad (11)$$

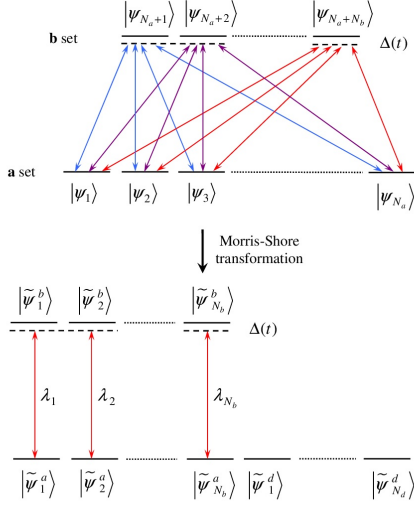


FIG. 1: Scheme of the Morris-Shore transformation, where a multistate system consisting of two coupled sets of degenerate levels is decomposed into a set of independent nondegenerate two-state systems and a set of decoupled states.

The structure of the MS Hamiltonian (11) shows that in the MS basis the dynamics is decomposed into sets of N_d decoupled single states, and N_b independent two-state systems $|\tilde{\psi}_n^a\rangle \leftrightarrow |\tilde{\psi}_n^b\rangle$ ($n = 1, 2, \dots, N_b$), each composed of an a state $|\tilde{\psi}_n^a\rangle$ and a b state $|\tilde{\psi}_n^b\rangle$, and driven by the RWA Hamiltonians,

$$\tilde{\mathbf{H}}_n(t) = \begin{bmatrix} 0 & \lambda_n \\ \lambda_n & \Delta(t) \end{bmatrix} \quad (n = 1, 2, \dots, N_b). \quad (12)$$

Each of these two-state Hamiltonians has the same detuning $\Delta(t)$, but they differ in the couplings λ_n . Each of the new a states $|\tilde{\psi}_n^a\rangle$ is the eigenstate of $\mathbf{V}\mathbf{V}^\dagger$ corresponding to the eigenvalue λ_n^2 , whereas each of the new b states $|\tilde{\psi}_n^b\rangle$ is the eigenstate of $\mathbf{V}^\dagger\mathbf{V}$, corresponding to the same eigenvalue λ_n^2 . The square root of this common eigenvalue, λ_n , represents the coupling in the respective independent MS two-state system $|\tilde{\psi}_n^a\rangle \leftrightarrow |\tilde{\psi}_n^b\rangle$ ($n = 1, 2, \dots, N_b$). The N_d zero eigenvalues of $\mathbf{V}\mathbf{V}^\dagger$ correspond to decoupled (dark) states in the a set (we assume throughout that $N_a \geq N_b$; therefore, dark states, if any, are in the a set). The dark states are decoupled from the dynamical evolution because they are driven by one-dimensional null Hamiltonians.

The MS decomposition described above allows us to reduce the degenerate two-level problem to a set of N_d nondegenerate two-state problems, wherein the detuning is unchanged while the couplings λ_n are combinations of the initial couplings between the a and b states and defined as the square roots of the eigenvalues of $\mathbf{V}^\dagger\mathbf{V}$.

From the vector form (4) of \mathbf{V} we obtain

$$\mathbf{V}\mathbf{V}^\dagger = \sum_{n=1}^{N_b} |\Omega_n\rangle \langle \Omega_n|, \quad (13a)$$

$$\mathbf{V}^\dagger\mathbf{V} = \begin{bmatrix} \langle \Omega_1 | \Omega_1 \rangle & \langle \Omega_1 | \Omega_2 \rangle & \cdots & \langle \Omega_1 | \Omega_{N_b} \rangle \\ \langle \Omega_2 | \Omega_1 \rangle & \langle \Omega_2 | \Omega_2 \rangle & \cdots & \langle \Omega_2 | \Omega_{N_b} \rangle \\ \vdots & \vdots & \ddots & \vdots \\ \langle \Omega_{N_b} | \Omega_1 \rangle & \langle \Omega_{N_b} | \Omega_2 \rangle & \cdots & \langle \Omega_{N_b} | \Omega_{N_b} \rangle \end{bmatrix} \quad (13b)$$

Note that $\mathbf{V}^\dagger\mathbf{V}$ is the Gramm matrix for the set of vectors $\{|\Omega_n\rangle\}_{n=1}^{N_b}$. Thus if all these vectors are linearly independent then $\det \mathbf{V}^\dagger\mathbf{V} \neq 0$ and all eigenvalues of $\mathbf{V}^\dagger\mathbf{V}$ are nonzero [13]; however, this assumption is unnecessary.

We assume that we can find the eigenvalues λ_n^2 ($n = 1, 2, \dots, N_b$) of the matrices (13a) and (13b), and the corresponding orthonormalized eigenvectors: the N_b coupled eigenstates $|\tilde{\psi}_n^a\rangle$ of $\mathbf{V}\mathbf{V}^\dagger$ and $|\tilde{\psi}_n^b\rangle$ of $\mathbf{V}^\dagger\mathbf{V}$, and the N_d decoupled eigenstates $|\tilde{\psi}_k^d\rangle$ of $\mathbf{V}\mathbf{V}^\dagger$. We use these eigenstates to construct the transformation matrices as

$$\mathbf{A} = \begin{bmatrix} \langle \tilde{\psi}_1^d | \\ \vdots \\ \langle \tilde{\psi}_{N_d}^d | \\ \langle \tilde{\psi}_1^a | \\ \vdots \\ \langle \tilde{\psi}_{N_b}^a | \end{bmatrix}, \quad \mathbf{B} = \begin{bmatrix} \langle \tilde{\psi}_1^b | \\ \vdots \\ \langle \tilde{\psi}_{N_b}^b | \end{bmatrix}. \quad (14)$$

B. The MS propagators

The propagator the independent MS two-state systems $|\tilde{\psi}_n^a\rangle \leftrightarrow |\tilde{\psi}_n^b\rangle$ ($n = 1, 2, \dots, N_b$), defined by

$$\begin{bmatrix} \tilde{C}_n^a(t_f) \\ \tilde{C}_n^b(t_f) \end{bmatrix} = \tilde{\mathbf{U}}_n(t_f, t_i) \begin{bmatrix} \tilde{C}_n^a(t_i) \\ \tilde{C}_n^b(t_i) \end{bmatrix}, \quad (15)$$

is unitary and can be parameterized in terms of the Cayley-Klein parameters

$$\tilde{\mathbf{U}}_n(t_f, t_i) = \begin{bmatrix} \alpha_n & -\beta_n^* \\ \beta_n & \alpha_n^* \end{bmatrix}. \quad (16)$$

Here, α_n and β_n are the Cayley-Klein parameters and they obey the relation

$$|\alpha|^2 + |\beta|^2 = 1. \quad (17)$$

C. The propagator in the diabatic basis

By taking into account the MS propagators (16) for the two-state MS systems, the ordering of the states, and the MS Hamiltonian (11), the propagator of the entire system in the MS basis is written as

$$\tilde{\mathbf{U}} = \begin{bmatrix} \mathbf{1}_{N_d} & \mathbf{0} \\ \mathbf{0} & \begin{matrix} \alpha_1 & 0 & \cdots & 0 & -\beta_1^* & 0 & \cdots & 0 \\ 0 & \alpha_2 & \cdots & 0 & 0 & -\beta_2^* & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \alpha_{N_b} & 0 & 0 & \cdots & -\beta_{N_b}^* \\ \beta_1 & 0 & \cdots & 0 & \alpha_1^* & 0 & \cdots & 0 \\ 0 & \beta_2 & \cdots & 0 & 0 & \alpha_2^* & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \beta_{N_b} & 0 & 0 & \cdots & \alpha_{N_b}^* \end{matrix} \end{bmatrix}. \quad (18)$$

By taking into account the completeness relations

$$\sum_{n=1}^{N_b} |\tilde{\psi}_n^a\rangle\langle\tilde{\psi}_n^a| + \sum_{k=1}^{N_d} |\tilde{\psi}_k^d\rangle\langle\tilde{\psi}_k^d| = \mathbf{1}_{N_a}, \quad (19a)$$

$$\sum_{n=1}^{N_b} |\tilde{\psi}_n^b\rangle\langle\tilde{\psi}_n^b| = \mathbf{1}_{N_b}, \quad (19b)$$

it is straightforward to show that the propagator in the original basis $\mathbf{U} = \mathbf{S}^\dagger \tilde{\mathbf{U}} \mathbf{S}$ reads

$$\mathbf{U} = \begin{bmatrix} \mathbf{1}_{N_a} + \sum_{n=1}^{N_b} (\alpha_n - 1) |\tilde{\psi}_n^a\rangle\langle\tilde{\psi}_n^a| & -\sum_{n=1}^{N_b} \beta_n^* |\tilde{\psi}_n^a\rangle\langle\tilde{\psi}_n^b| \\ \sum_{n=1}^{N_b} \beta_n |\tilde{\psi}_n^b\rangle\langle\tilde{\psi}_n^a| & \sum_{n=1}^{N_b} \alpha_n^* |\tilde{\psi}_n^b\rangle\langle\tilde{\psi}_n^b| \end{bmatrix} \quad (20)$$

Note that the propagator does not depend on the decoupled states $|\tilde{\psi}_k^d\rangle$ ($k = 1, 2, \dots, N_d$), which are excluded by using Eq. (19a). This has to be expected because, owing to their degeneracy, the choice of the decoupled states is not unique, since any superposition of them is also a zero-eigenvalue eigenstate of $\mathbf{V}\mathbf{V}^\dagger$. Because the dynamics in the original basis must not depend on such arbitrariness, the propagator \mathbf{U} must not depend on the decoupled states at all.

IV. MS WITH NONDEGENERATE(UNEQUAL) DETUNINGS

We

$$\mathbf{H}(t) = \begin{bmatrix} \mathbf{D}_1(t)_{N_a \times N_a} & \mathbf{V}(t)_{N_a \times N_b} \\ \mathbf{V}^\dagger(t)_{N_b \times N_a} & (\boldsymbol{\Delta}(t) + \mathbf{D}_2(t))_{N_b \times N_b} \end{bmatrix} = \mathbf{H}_0(t) + \mathbf{D}(t). \quad (21)$$

where $\mathbf{D}(t)_{(N_a+N_b) \times (N_a+N_b)}$ is diagonal matrix, given by

$$\mathbf{D}(t) = \begin{bmatrix} \mathbf{D}_1(t)_{N_a \times N_a} & \mathbf{0} \\ \mathbf{0} & \mathbf{D}_2(t)_{N_b \times N_b} \end{bmatrix}$$

$$\mathbf{D}_1(t) = \text{diag} [d_1^1(t), d_2^1(t), \dots, d_{N_a}^1(t)], \quad (22)$$

$$\mathbf{D}_2(t) = \text{diag} [d_1^2(t), d_2^2(t), \dots, d_{N_b}^2(t)], \quad (23)$$

The main difficulty for finding exact solution is that in the MS basis we have

$$\begin{aligned} \tilde{\mathbf{H}}(t) &= \mathbf{S} \mathbf{H}(t) \mathbf{S}^\dagger = \begin{bmatrix} \mathbf{A} \mathbf{D}_1(t) \mathbf{A}^\dagger & \tilde{\mathbf{V}} \\ \tilde{\mathbf{V}}^\dagger & \boldsymbol{\Delta}(t) + \mathbf{B} \mathbf{D}_2(t) \mathbf{B}^\dagger \end{bmatrix} \\ &= \mathbf{S} (\mathbf{H}_0(t) + \mathbf{D}(t)) \mathbf{S}^\dagger = \mathbf{H}_0^{MS}(t) + \mathbf{S} \mathbf{D}(t) \mathbf{S}^\dagger \end{aligned}$$

The matrices $\mathbf{A} \mathbf{D}_1(t) \mathbf{A}^\dagger \neq \mathbf{D}_1(t)$ and $\mathbf{B} \mathbf{D}_2(t) \mathbf{B}^\dagger \neq \mathbf{D}_2(t)$ are not diagonal. This simply means in the MS basis the dynamics cannot be decomposed into sets of N_d decoupled single states, and N_b independent two-state systems. Hereafter for convenience we will assume $\mathbf{D}_1(t) = 0$ and the notation $\mathbf{D}_2(t) = \mathbf{D}(t)$ will be used.

Instead of exact factorization we look at solution that can be expressed in perturbative form. The Hamiltonian in a block-matrix form is given by $\mathbf{H}(t) = \mathbf{H}_0(t) + \epsilon \mathbf{D}(t)$. We will look for perturbative solution for the MS problem and for this reason we will have the following definition for the transformation matrix \mathbf{S} .

$$\mathbf{S} = \mathbf{S}_0 + \epsilon \mathbf{S}_1 + \epsilon^2 \mathbf{S}_2 + \dots \quad (24)$$

where the additional condition $\dot{\mathbf{S}} = \mathbf{0}$ is imposed. This simply means that all terms from the series expansion in Eq.(24) are constant matrices. Also in reason to simplify the calculations and without loss of generality the \mathbf{S}_i matrices have the block-diagonal form, similar to (7)

$$\mathbf{S}_i = \begin{bmatrix} \mathbf{A}_i & \mathbf{0} \\ \mathbf{0} & \mathbf{B}_i \end{bmatrix}. \quad (25)$$

According to the Eq.(6) and using the series expansion for the transformation matrix \mathbf{S} given in Eq.(24) we will obtain

$$\begin{aligned} \mathbf{C}(t) &= \mathbf{S}^\dagger \tilde{\mathbf{C}}(t) \\ &= (\mathbf{S}_0 + \epsilon \mathbf{S}_1 + \epsilon^2 \mathbf{S}_2 + \dots) \tilde{\mathbf{C}}(t) \\ &= \tilde{\mathbf{C}}_0(t) + \epsilon \tilde{\mathbf{C}}_1(t) + \epsilon^2 \tilde{\mathbf{C}}_2(t) + \dots \end{aligned} \quad (26)$$

From the expression above is clear the meaning of the $\tilde{\mathbf{C}}_i(t)$ state vectors.

Using the series expansion for the transformation matrix \mathbf{S} we will obtain the following expression for the Hamiltonian in the new basis

$$\begin{aligned} \tilde{\mathbf{H}}(t) &= \mathbf{S} \mathbf{H}(t) \mathbf{S}^\dagger = \\ &= \left(\sum_{i=0} \epsilon^i \mathbf{S}_i \right) [\mathbf{H}_0(t) + \epsilon \mathbf{D}(t)] \left(\sum_{i=0} \epsilon^i \mathbf{S}_i^\dagger \right) \\ &= \mathbf{S}_0 \mathbf{H}_0 \mathbf{S}_0^\dagger + \epsilon [\mathbf{S}_0 \mathbf{D} \mathbf{S}_0^\dagger + \mathbf{S}_1 \mathbf{H}_0 \mathbf{S}_0^\dagger + \mathbf{S}_0 \mathbf{H}_0 \mathbf{S}_1^\dagger] \\ &\quad + \epsilon^2 [\mathbf{S}_0 \mathbf{D} \mathbf{S}_1^\dagger + \mathbf{S}_1 \mathbf{D} \mathbf{S}_0^\dagger + \\ &\quad \mathbf{S}_0 \mathbf{H}_0 \mathbf{S}_2^\dagger + \mathbf{S}_1 \mathbf{H}_0 \mathbf{S}_1^\dagger + \mathbf{S}_2 \mathbf{H}_0 \mathbf{S}_2^\dagger] + O(\epsilon^3) \end{aligned} \quad (27)$$

In the above expression for the Hamiltonian only the first two terms in the power expansion of ε are given, but the reader can easily take more terms for this perturbative expansion.

Let the matrix \mathbf{S}_0 be chosen such that

$$\mathbf{S}_0 \mathbf{H}_0 \mathbf{S}_0^\dagger = \mathbf{H}_0^{MS}(t), \text{ } \mathbf{S}_0\text{- definition} \quad (29)$$

and $\mathbf{H}_0^{MS}(t)$ has the form

$$\mathbf{H}_0^{MS}(t) = \begin{bmatrix} \mathbf{0} & \tilde{\mathbf{V}} \\ \tilde{\mathbf{V}}^\dagger & \mathbf{\Delta} \end{bmatrix},$$

where $\tilde{\mathbf{V}}$ is given by

$$\tilde{\mathbf{V}} = \begin{bmatrix} \mathbf{0}_{N_d \times N_b} \\ \mathbf{\Lambda}_{N_b \times N_b} \end{bmatrix}.$$

The square matrix $\mathbf{\Lambda}$ is diagonal,

$$\mathbf{\Lambda}(t) = \text{diag}[\lambda_1(t), \lambda_2(t), \dots, \lambda_{N_b}(t)]$$

This choice for the matrix \mathbf{S}_0 corresponds to neglecting the perturbative term $\varepsilon \mathbf{D}(t)$.

Having in mind that the matrix \mathbf{S}_0 is computed, using Eq.(27) we can continue and take the corrections up to first order of ε by choosing matrix \mathbf{S}_1 from the equation

$$\mathbf{S}_1 \mathbf{H}_0(t) \mathbf{S}_0^\dagger + \mathbf{S}_0 \mathbf{H}_0(t) \mathbf{S}_1^\dagger + \mathbf{S}_0 \mathbf{D}(t) \mathbf{S}_0^\dagger = 0, \text{ } \mathbf{S}_1\text{- definition} \quad (30)$$

Using the block-matrix forms for $\mathbf{H}_0(t)$, $\mathbf{D}(t)$, \mathbf{S}_0 and \mathbf{S}_1 matrices and writing the third term to the rhs of the equation the above expression reads

$$\begin{bmatrix} \mathbf{0} & \mathbf{A}_1 \mathbf{V} \mathbf{B}_0^\dagger \\ \mathbf{B}_1 \mathbf{V}^\dagger \mathbf{A}_0^\dagger & \mathbf{B}_1 \mathbf{\Delta} \mathbf{B}_0^\dagger \end{bmatrix} + \begin{bmatrix} \mathbf{0} & \mathbf{A}_0 \mathbf{V} \mathbf{B}_1^\dagger \\ \mathbf{B}_0 \mathbf{V}^\dagger \mathbf{A}_1^\dagger & \mathbf{B}_0 \mathbf{\Delta} \mathbf{B}_1^\dagger \end{bmatrix} = - \begin{bmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{B}_0 \mathbf{D} \mathbf{B}_1^\dagger \end{bmatrix} \quad (31)$$

The above block-matrix equation is equivalent to the system of matrix equations

$$\mathbf{A}_1 \mathbf{V} \mathbf{B}_0^\dagger + \mathbf{A}_0 \mathbf{V} \mathbf{B}_1^\dagger = 0 \quad (32)$$

$$\mathbf{B}_1 \mathbf{V}^\dagger \mathbf{A}_0^\dagger + \mathbf{B}_0 \mathbf{V}^\dagger \mathbf{A}_1^\dagger = 0 \quad (33)$$

$$\mathbf{B}_1 \mathbf{\Delta} \mathbf{B}_0^\dagger + \mathbf{B}_0 \mathbf{\Delta} \mathbf{B}_1^\dagger = -\mathbf{B}_0 \mathbf{D} \mathbf{B}_1^\dagger \quad (34)$$

The first and the second equation from the above system are equivalent under the hermitian conjugate operation. We want to note that from Eq.(29) the \mathbf{A}_0 and \mathbf{B}_0 matrices are known. Hence the system of equation given by Eq.(33) and Eq.(34), can be solved for \mathbf{A}_1 and \mathbf{B}_1 matrices, which defines the transformation matrix \mathbf{S}_1 .

Before we continue with \mathbf{S}_2 matrix computation we would like to simplify the matrix equation Eq.(34). Remember that the matrix $\mathbf{\Delta}(t) = \delta(t) \mathbf{1}_{N_b}$, one can use the commutation relations $\mathbf{B}_1 \mathbf{\Delta} = \mathbf{\Delta} \mathbf{B}_1$ and $\mathbf{B}_0 \mathbf{\Delta} = \mathbf{\Delta} \mathbf{B}_0$, and the unitary properties \mathbf{B}_0 and \mathbf{B}_1 to transform Eq.(34).

$$\begin{aligned} \mathbf{\Delta} (\mathbf{B}_1 \mathbf{B}_0^\dagger + \mathbf{B}_0 \mathbf{B}_1^\dagger) &= -\mathbf{B}_0 \mathbf{D} \mathbf{B}_1^\dagger \\ (\mathbf{B}_1 \mathbf{B}_0^\dagger + \mathbf{B}_0 \mathbf{B}_1^\dagger) &= -\mathbf{B}_0 \mathbf{\Delta}^{-1} \mathbf{D} \mathbf{B}_1^\dagger \end{aligned}$$

Using the unitary properties for the matrices \mathbf{B}_0 and \mathbf{B}_1 for Eq.(34) the system of matrix equation, which defines the transformation matrix \mathbf{S}_1 can be written in the form

$$\begin{aligned} \mathbf{A}_1 \mathbf{V} \mathbf{B}_0^\dagger + \mathbf{A}_0 \mathbf{V} \mathbf{B}_1^\dagger &= 0 \\ \mathbf{B}_0^\dagger \mathbf{B}_1 + \mathbf{B}_1^\dagger \mathbf{B}_0 &= -\mathbf{\Delta}^{-1} \mathbf{D} \end{aligned} \quad (35)$$

The algorithm for deriving expression for the \mathbf{S}_2 matrix is similar to this for \mathbf{S}_1 matrix. Having in mind that the matrices \mathbf{S}_0 and \mathbf{S}_1 are computed, we can continue and take the corrections up to ε^2 by choosing matrix \mathbf{S}_2 from the equation

$$\begin{aligned} \mathbf{S}_0 \mathbf{D}(t) \mathbf{S}_1^\dagger + \mathbf{S}_1 \mathbf{D}(t) \mathbf{S}_0^\dagger + \\ \mathbf{S}_0 \mathbf{H}_0(t) \mathbf{S}_2^\dagger + \mathbf{S}_1 \mathbf{H}_0(t) \mathbf{S}_1^\dagger + \mathbf{S}_2 \mathbf{H}_0(t) \mathbf{S}_2^\dagger &= 0, \text{ } \mathbf{S}_2\text{- definition} \end{aligned} \quad (36)$$

By similar methods to the presented for the matrix \mathbf{S}_2 , one can derive the higher order correction i.e., find matrices \mathbf{S}_3 , \mathbf{S}_4 , .. etc.

V. EXAMPLE

VI. THE CASE $N_b = 2$

Above, we described the method for deriving solution to the generalized MS problem, where the evolution of a two-level quantum system is subjected to pulsed external field and in the general case the ground and excited levels have arbitrary level of degeneracy, N_a and N_b respectively. In this section, we will illustrate these results with a specific example: when the \mathbf{b} set consists of two

degenerate states, i.e. $N_b = 2$. This case is interesting, because of the possible realizations of our model in different real physical systems. Moreover, this special case has an exact analytical solution, which is very rare for multidimensional systems.

A. General case

We label the ground states $|\psi_i\rangle$ ($i = 1, 2, \dots, N$), where N is arbitrary and we denote the two excited states $|\psi'\rangle$ and $|\psi''\rangle$. In this case the interaction operator \mathbf{V} (3) has N columns and two rows, and its explicit form is given by

$$\mathbf{V} = \begin{bmatrix} \Omega'_1 & \Omega''_1 \\ \Omega'_2 & \Omega''_2 \\ \vdots & \vdots \\ \Omega'_N & \Omega''_N \end{bmatrix} = [|\Omega'\rangle, |\Omega''\rangle], \quad (37)$$

where $|\Omega'\rangle$ and $|\Omega''\rangle$ are N -dimensional vectors comprising the couplings between the ground states and the corresponding excited state

$$|\Omega'\rangle = \begin{bmatrix} \Omega'_1 \\ \Omega'_2 \\ \vdots \\ \Omega'_N \end{bmatrix}, \quad |\Omega''\rangle = \begin{bmatrix} \Omega''_1 \\ \Omega''_2 \\ \vdots \\ \Omega''_N \end{bmatrix}. \quad (38)$$

We write the product of $\mathbf{V}^\dagger \mathbf{V}$

$$\mathbf{V}^\dagger \mathbf{V} = \begin{bmatrix} |\Omega'|^2 & \langle \Omega' | \Omega'' \rangle \\ \langle \Omega' | \Omega'' \rangle^* & |\Omega''|^2 \end{bmatrix}, \quad (39)$$

with eigenvalues,

$$\lambda_\pm^2 = \frac{1}{2} [|\Omega'|^2 + |\Omega''|^2 \pm \sqrt{D}], \quad (40)$$

where D denotes

$$D = (|\Omega'|^2 - |\Omega''|^2)^2 + 4|\langle \Omega' | \Omega'' \rangle|^2 = \frac{(|\Omega'|^2 - |\Omega''|^2)^2}{\cos^2 2\theta}, \quad (41)$$

and we have introduced the following parameterizations,

$$\frac{2|\langle \Omega' | \Omega'' \rangle|}{|\Omega''|^2 - |\Omega'|^2} = \tan 2\theta, \quad (0 < 2\theta < \pi) \quad (42)$$

$$\arg \langle \Omega' | \Omega'' \rangle = \sigma. \quad (43)$$

It is straightforward to obtain the eigenstates $|\tilde{\psi}_+^b\rangle$ and $|\tilde{\psi}_-^b\rangle$, corresponding to λ_+^2 and λ_-^2 , which represent the excited states in the MS two-state systems,

$$|\tilde{\psi}_+^b\rangle = \frac{1}{n_+} \begin{bmatrix} \langle \Omega' | \Omega'' \rangle \\ \lambda_+ - |\Omega'|^2 \end{bmatrix} = \begin{bmatrix} e^{i\sigma} \sin \theta \\ \cos \theta \end{bmatrix}, \quad (44a)$$

$$|\tilde{\psi}_-^b\rangle = \frac{1}{n_-} \begin{bmatrix} \langle \Omega' | \Omega'' \rangle \\ \lambda_- - |\Omega'|^2 \end{bmatrix} = \begin{bmatrix} e^{i\sigma} \cos \theta \\ -\sin \theta \end{bmatrix}, \quad (44b)$$

with n_+ and n_- normalization factors.

The next step is to find the Householder vectors $|\tilde{\psi}_+^a\rangle$ and $|\tilde{\psi}_-^a\rangle$, which are the ground states in the MS two-state systems and define the propagator \mathbf{U}_a . They are the eigenstates of the N -dimensional matrix,

$$\mathbf{V}\mathbf{V}^\dagger = |\Omega'\rangle \langle \Omega'| + |\Omega''\rangle \langle \Omega''|, \quad (45)$$

corresponding to the same non-zero eigenvalues (40). We construct them as superpositions of the interaction vectors with coefficients α'_+ , α''_+ , α'_- and α''_- ,

$$|\tilde{\psi}_\pm^a\rangle = \alpha'_\pm |\Omega'\rangle + \alpha''_\pm |\Omega''\rangle. \quad (46)$$

We determine these coefficients from the eigenvalue equations,

$$\mathbf{V}\mathbf{V}^\dagger (\alpha'_\pm |\Omega'\rangle + \alpha''_\pm |\Omega''\rangle) = \lambda_\pm (\alpha'_\pm |\Omega'\rangle + \alpha''_\pm |\Omega''\rangle), \quad (47)$$

and from the normalization conditions,

$$\langle \tilde{\psi}_\pm^a | \tilde{\psi}_\pm^a \rangle = 1. \quad (48)$$

As a result we obtain

$$|\tilde{\psi}_+^a\rangle = \frac{1}{\lambda_+} (\cos \theta |\Omega'\rangle + e^{-i\sigma} \sin \theta |\Omega''\rangle), \quad (49)$$

$$|\tilde{\psi}_-^a\rangle = \frac{1}{\lambda_-} (\sin \theta |\Omega'\rangle - e^{-i\sigma} \cos \theta |\Omega''\rangle). \quad (50)$$

The results given in equations Eq.(44a), Eq.(44b), Eq.(49) and Eq.(50) define the block-structure of the \mathbf{S}_0 matrix via Eq.(25). Using Eq.(44a), Eq.(44b) the \mathbf{B}_0 matrix reads

$$\mathbf{B}_0 = \begin{bmatrix} e^{-i\sigma_0} \sin \theta_0 & \cos \theta_0 \\ e^{-i\sigma_0} \cos \theta_0 & -\sin \theta_0 \end{bmatrix} \quad (51)$$

We will look for solution for the matrix \mathbf{B}_1 , and without loss of generality one can assume that \mathbf{B}_1 possesses the same functional form as \mathbf{B}_0

$$\mathbf{B}_1 = \begin{bmatrix} e^{-i\sigma_1} \sin \theta_1 & \cos \theta_1 \\ e^{-i\sigma_1} \cos \theta_1 & -\sin \theta_1 \end{bmatrix}, \quad (52)$$

where the matrix parameters σ_1 and θ_1 need to be derived. Using the matrix system of equations given by Eq.(35) the solutions for \mathbf{A}_1 and \mathbf{B}_1 can be derived.

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